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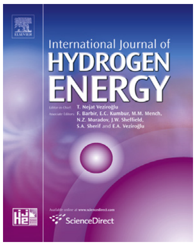

Simulation of exhaust gas reforming of propane in a heat exchange integrated microchannel reactor

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ABSTRACT

Exhaust gas reforming of propane to a hydrogen-rich mixture in a single, heat-exchange integrated, adiabatic, catalytic microchannel is modeled and simulated at different exhaust gas compositions from conventional gasoline and diesel fueled engines. Propane is considered as the model hydrocarbon for the complex fuels such as gasoline and diesel. The single microchannel is considered to be the characteristic unit of the catalytic exhaust gas reformer involving identical channels located parallel to each other. Steady-state simulations, carried out by the finite volume method, involve parametric variations of the total feed flow rate, and the amounts of propane and steam injected externally into the exhaust gas (reformer feed) stream. The results show that effective heat transfer and uniform temperature distribution, which are critical for the successful operation of the exhaust gas reformer, can be obtained in the microchannel configuration even at low gas hourly space velocities (GHSVs) at which the conventional packed-bed reformers usually lead to remarkable hot-spot formation. Production of H_2 and CO is favored by the addition of higher amounts of propane and steam into the reformer feed. Increasing the total feed flow rate, hence the GHSV is found to improve heat distribution along the microchannel at the expense of reduced product yields due to insufficient contact time.

1. Introduction

Exhaust gas reforming (EGR) is a novel fuel conversion technique that is proposed to improve the efficiency and reduce the emissions of conventional gasoline and diesel fuel based engines [1]. In this system, a compact, catalytic reformer is placed into the exhaust gas recirculation loop to react the hot exhaust gas stream with externally injected fuel and steam for producing a hydrogen-rich mixture. The hydrogen-rich effluent of the reformer is then fed back to the engine to improve combustion efficiency and reduce undesired emissions [1]. Energy required to sustain EGR is supplied by the sensible heat of the exhaust gas stream generated in the engine and by the catalytic oxidation of part of the fuel that is added to the reformer. Remaining part of the fuel is then reformed into a hydrogen-rich mixture by steam that already exists in the exhaust stream as a product of engine operation and is also supplied externally [1]. Success of the EGR depends on catalyst type and reactor configuration. Pt and Rh-based catalysts are known as the best options for driving the main reactions of EGR, namely total oxidation (TOX) and steam reforming (SR) of the injected fuel, respectively [2,3]:

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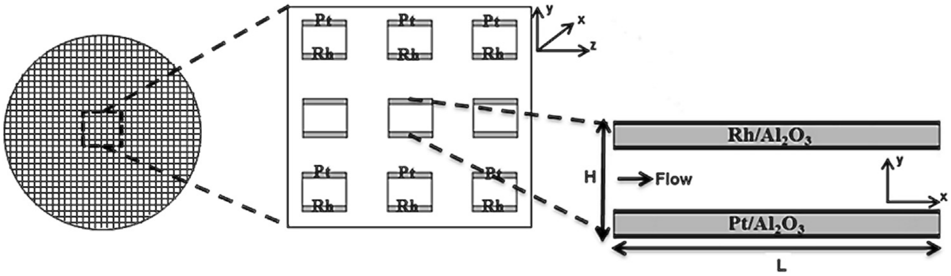

Fig. 1 – Microchannel geometry.

$$\text{TOX: } C_3H_8 + 5O_2 \rightarrow 3CO_2 + 4H_2O,\ \Delta H^o = -2044 \text{ kJ/mol} \quad (1)$$

$$\text{SR: } C_3H_8 + 3H_2O \Leftrightarrow 3CO + 7H_2,\ \Delta H^o = 498 \text{ kJ/mol} \quad (2)$$

The reactions are given for propane, which is reported as the surrogate for the conventional hydrocarbon fuels such as gasoline and diesel [4]. In addition to these key reactions, product distribution in EGR is also affected by the water–gas shift reaction (WGS):

$$\text{WGS: } CO + H_2O \Leftrightarrow CO_2 + H_2,\ \Delta H^o = -41.2 \text{ kJ/mol} \quad (3)$$

It is reported that the exothermic TOX and endothermic SR occur consecutively during EGR [4,5]. At this point, the reactor configuration becomes very critical, as it has significant impact on the rate of transfer of the heat supplied to the reformer and generated by TOX to endothermic SR [4–6]. Current studies consider the use of either packed bed or monolith reactors [6,7]. Temperature profile of EGR in a packed-bed type reformer is typically characterized by a notable hot spot formation, which is an indication of uneven axial heat distribution [6,7]. This situation, caused by the inherently weak heat transfer properties of packed beds, can be enhanced by using a wall-coated monolith reactor in which the difference between maximum and reactor exit temperatures is much less than that observed in a packed bed configuration. In monolith reactors containing precious metal catalysts, the catalyst activity is reported to be very high and close to equilibrium in the hot zone, provided that the mass transfer limitations are overcome. The extent of these contributions is determined by the position and size of the temperature peak in the catalyst bed, both of which depend strictly on the reactor configuration [7].

Microchannel technology involves the use of structured flow paths or channels with hydraulic diameters in the submillimeter range. Small channel diameters result in very high surface area-to-volume ratios (ca. 1–5 × 10^4 m^2 m^{-3}), while this ratio is around 1 × 10^2 m^2 m^{-3} and rarely exceeds 1 × 10^3 m^2 m^{-3} in a conventional reactor [8,9]. The net result is significant volume reduction (up to ca. 90%), which leads to notable improvements in energy efficiency [10]. Microchannel reactors operate under laminar flow conditions which make them advantageous in terms of enhanced heat transfer characteristics [9]. Since the Nusselt number is constant and heat transfer coefficient is inversely proportional to the channel diameter in microchannels, the values can become one order of magnitude higher than those involved in the traditional units [9]. Compact and metallic nature of the microchannel units also favor better heat transfer along the catalyst layer, which is utilized efficiently during the reactions without the formation of any hot or cold spots [11]. These properties make microchannel reactors promising for use in EGR in which heat transfer is critical: energy supplied by the sensible heat of the engine exhaust stream and by the exothermal heat of oxidation of the part of the fuel injected into the reformer should be effectively transferred to the endothermic reforming of the remaining fuel to favor synthesis of a hydrogen-rich product [12].

The combination of Reactions (1)–(3) is also known as oxidative steam reforming (OSR). Use of microchannel units in reaction engineering is a novel concept and offers many advantages in applications such as OSR, which combines the heat effects of TOX and SR by taking fuel, steam and air as reactants [13]. Propane OSR over Rh-impregnated metallic microchannel reactors was studied by Aartun et al. [14,15]. Temperature profiles obtained along the reactor axis proved that the use of metallic microchannel reactor minimized the temperature gradients resulting from the exothermic nature of the OSR. The authors also investigated the effect of total flow rate and reported that at residence times less than 10 ms, propane conversion and selectivities to H_2 and CO were increased. Moreover, production of undesired by-products such as methane and ethane was reduced. Kaisare et al. [16] developed a pseudo-two-dimensional model to understand the stability and performance of Pt-catalyzed microreactors for propane combustion. Thermal conductivity of the reactor solid structure and inlet velocity were demonstrated to have strong impacts on heat and mass transfer rates along the microreactor. Pennemann et al. [17] studied conversion of propane with steam and oxygen in a stainless steel microchannel reactor involving washcoated forms of γ-Al_2O_3-supported Rh, Pd and Pt catalysts. Strong dependence of propane conversion on the catalyst type was demonstrated. It was also reported that complete propane conversion and near-equilibrium selectivities towards H_2 and CO were observed over the Rh-based catalysts. Pagani et al. [18] conducted a detailed kinetic analysis of TOX and SR of propane over 2%Rh/δ-Al_2O_3 in an annular microreactor. The authors concluded that H_2 and CO were produced through consecutive steps of TOX and SR of propane. The rates of propane oxidation and reforming were found to be proportional to the fuel concentration with first-order dependence.